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We analyze an interferometric complementarity between one- and two-particle interference in the general case: $V_i^2 + V_{12}^2 \leq 1$ ($i = 1, 2$), and further examine the relation among one-particle interference visibility V_i, two-particle interference visibility V_{12} and the predication P_i of the path of a single particle. An equality $V_i^2+V_{12}^2+P_i^2 = 1$ ($i = 1, 2$) is achieved for any pure two-particle source, which implies the condition of the complementarity relation to reach the upper bound and its relation to another interferometric complementarity between path information and interference visibility of a single particle. Meanwhile, the relationships of the complementarities and the entanglement E of the composite system are also investigated. Using nuclear magnetic resonance techniques, the two-particle interferometric complementarity was experimentally tested with the ensemble-averaged spin states, including two extreme cases and an intermediate case.

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I. INTRODUCTION

Quantum superposition and its resulting interference are arguably the most fundamental effects in quantum mechanics, which leads to the concept of complementarity. In 1927, Bohr [1] first reviewed this subject, claimed that the wave- and particle-like behaviors of a quantum mechanical object are mutually exclusive in a single experiment and expressed it as complementarity. So Bohr complementarity is often superficially identified with the 'wave-particle duality' [2,3] which emphasizes two extreme cases that each experiment must be described in terms of either waves or particles, as explained in the textbook. Theoretical investigations about the intermediate situations in which particle and wave aspects coexist have led to some quantitative statements about wave-particle duality [4–10], some of which are expressed as inequalities $V^2 + P^2 \leq 1$ or $V^2 + D^2 \leq 1$ about the complementarity between 'which-way' (WW) information of a particle: the predictability P and the distinguishability D of the path, and the visibility V of interference fringes [8–10]. Bohr complementarity is often illustrated by means of interferometers and has been experimentally investigated in one-particle interferometer with individual particles, including photons [11–13], electrons [14], neutrons [15–17], atoms [18–20] and nuclear spins in a bulk ensemble with nuclear magnetic resonance (NMR) techniques [21,22].

Complementarity, however, is a more general concept which states that quantum systems possess properties that are equally real but mutually exclusive. Naturally, the techniques of standard one-particle interferometry have been extended to two-particle interferometry by employing correlated two-particle systems. The two-particle interference phenomenon was first observed by Ghosh and Mandel [23] using photon pair produced by parametric down-conversion, where the nonclassical effects associated with Einstein-Podolsky-Rosen (EPR) states were exhibited. Since that, two-particle interferometry has been intensively studied theoretically and experimentally [24]. Whereafter, a fact between one- and two-particle interference was noticed by Horne and Zeilinger [25], i.e., when the two-particle visibility is unity, the one-particle visibility is zero, and vice versa. The visibilities of the one- and two-particle interference are described as a pair of complementary quantities. A systematic investigation of intermediate cases was carried out by Jaeger et al. [26], who showed that in a large family of states $|\Theta\rangle$ an interferometric complementarity holds between one-particle visibility V_i and two-particle visibility V_{12}:

$$V_i^2 + V_{12}^2 \leq 1, (i = 1, 2) \tag{1}$$

where the upper bound is saturated for a special family of states while most of the cases are limited in the inequality (1). However, the condition of achieving the upper bound has not yet been pointed out. The complementarity relation of one- and two-photon interference has been experimentally demonstrated in a Young's double-slit experiment by

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Abouraddy et al. [27]. Except this experiment, we have not still found other experimental versions in the intermediate regime. Moreover, inequality (1) can be strengthened to an equality with a suitable extension of operations on the pair of particles [8], but this conclusion is based on the different parameters of experimental configuration, that is, one can not observe such interference fringes to obtain V_i and V_{12} that satisfy the equality in practice [8].

Then, what is the condition of this complementarity being of the form of an equality $V_i^2 + V_{12}^2 = 1$? And what is the reason why the complementarity relation is restricted in the inequality (1)? In this paper, we analyze the one- and two-particle interference complementarity in the general case and examine the relation among V_i, V_{12} and the predication of the path P_i to obtain an equality: $V_i^2+V_{12}^2+P_i^2 = 1, \quad (i = 1,2)$ for any pure two-particle source, which gives the answers of the two questions above, and, in fact, embodies two interferometric complementarity relations whose relations are discussed. Furthermore, we also clarify the relationships of V_{12}, the distinguishability of the path D_i and the entanglement E of the composite system which show an important view that this complementarity can arise due to the correlations. Using NMR techniques detailed in our works [21,22], the one- and two-particle complementarity relation of $V_i^2 + V_{12}^2 = 1$ with $P_i = 0$ has been experimentally tested with the ensemble-averaged spin states of ^{13}C and ^{1}H nuclei in ^{13}C-labelled chloroform $^{13}CHCl_3$, including two extreme cases: perfect two-particle interference fringe and no one-particle interference fringe for an entangled state and the opposite for a product state, and an intermediate case of a special family of states, by measuring both single and joint probability densities of spin states.

II. COMPLEMENTARITY IN TWO EXTREME CASES

We briefly review some dinfinitions and two-particle interferometric complementarity in two extreme cases. A schematic arrangement for a two-particle interferometer with variable phase shifters is given in Fig. 1 [26]. The source S emits a pair of particles 1+2, one of which propagates along the path A and/or A$'$, through a variable phase shifter ϕ_1 impinging on an ideal symmetric beam splitter BS_1, and is then registered in either beam K_1 or L_1, while on the other side there is the analogous process on the other (see Fig. 1). The locution "and/or" refers in a brief way to a quantum-mechanic superposition, the details of which are specified by the state of particles 1+2. Assuming that the pair of particles 1+2 are ideally two-level or spin-1/2 quantum entities, the Hilbert space H_1 associated with particle 1 is spanned by vectors $|A\rangle$ and $|A'\rangle$ representing states of propagation in the paths A and A$'$, and the Hilbert space H_2 associated with particle 2 by analogous vectors $|B\rangle$ and $|B'\rangle$. In the same way, the space of the output states is the subspaces spanned by the vectors $|K_1\rangle$ and $|L_1\rangle$ for particle 1 and by $|K_2\rangle$ and $|L_2\rangle$ for particle 2.

According to the standard definition of visibility $V = (I_{max} - I_{min})/(I_{max} + I_{min})$, the usual definition for single-particle fringe visibility is

$$V_i = \frac{[p(K_i)]_{\max} - [p(K_i)]_{\min}}{[p(K_i)]_{\max} + [p(K_i)]_{\min}}, \qquad (i = 1,2), \tag{2}$$

whereas for two-particle fringe visibility the "corrected" definition [8,26] is used:

$$V_{12} = \frac{[\overline{p}(K_1K_2)]_{\max} - [\overline{p}(K_1K_2)]_{\min}}{[\overline{p}(K_1K_2)]_{\max} + [\overline{p}(K_1K_2)]_{\min}}, \tag{3}$$

where the "corrected" joint probability $\overline{p}(K_1K_2) = p(K_1K_2) - p(K_1)p(K_2) + \frac{1}{4}$ to assure the reasonableness of the visibility V_{12} [8,26], and $p(K_i)$ and $p(K_1K_2)$ denote the probabilities of single and joint detections, respectively. These visibilities can be obtained by observing the variations of both single and joint probabilities as functions of the phases ϕ_1 and ϕ_2. For simplicity, our experiments take the case of $|A\rangle = |B\rangle = |K_1\rangle = |K_2\rangle = |\uparrow\rangle = \begin{pmatrix}1\\0\end{pmatrix}$ and $|A'\rangle = |B'\rangle = |L_1\rangle = |L_2\rangle = |\downarrow\rangle = \begin{pmatrix}0\\1\end{pmatrix}$ representing the single-particle states with spin-up and spin-down, respectively.

Consider two extreme cases [26]: when particles 1+2 is prepared in an entangled state

$$|\Psi\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\rangle_1|\uparrow\rangle_2 + |\downarrow\rangle_1|\downarrow\rangle_2\right), \tag{4}$$

or in a product state

$$|\Phi\rangle = \left[\frac{1}{\sqrt{2}}\left(|\uparrow\rangle_1 + |\downarrow\rangle_1\right)\right] \otimes \left[\frac{1}{\sqrt{2}}\left(|\uparrow\rangle_2 + |\downarrow\rangle_2\right)\right]. \tag{5}$$

The joint action of variable phase shifters and symmetric beam splitters can be described by a unitary operation

$$U(\phi_1,\phi_2)=U_1(\phi_1)\otimes U_2(\phi_2) \tag{6}$$

where

$$U_i(\phi_i)=\frac{1}{\sqrt{2}}\begin{pmatrix}1 & e^{i\phi_i}\\ -e^{-i\phi_i} & 1\end{pmatrix} \tag{7}$$

and subscript i represents particle i. $U(\phi_1,\phi_2)$ is the unitary mapping from the space of input states of the pair of particles 1+2 into the space of output states. It is easy to show that the joint and single measurement probabilities in the space of output states are: for the $|\Psi\rangle$ state of Eq.(4),

$$\begin{array}{l} p\left(|\uparrow\rangle_1|\uparrow\rangle_2\right)=p\left(|\downarrow\rangle_1|\downarrow\rangle_2\right)=\frac{1}{4}\left[1+\cos(\phi_1+\phi_2)\right]\\ p\left(|\uparrow\rangle_1|\downarrow\rangle_2\right)=p\left(|\downarrow\rangle_1|\uparrow\rangle_2\right)=\frac{1}{4}\left[1-\cos(\phi_1+\phi_2)\right]\\ p\left(|\uparrow\rangle_1\right)=p\left(|\downarrow\rangle_1\right)=p\left(|\uparrow\rangle_2\right)=p\left(|\downarrow\rangle_2\right)=\frac{1}{2}\end{array} \tag{8}$$

and for the $|\Phi\rangle$ state of Eq.(5),

$$\begin{array}{l} p\left(|x\rangle_1|y\rangle_2\right)=p\left(|x\rangle_1\right)p\left(|y\rangle_2\right)\\ p\left(|\uparrow\rangle_i\right)=\frac{1}{2}\left(1+\cos\phi_i\right)\\ p\left(|\downarrow\rangle_i\right)=\frac{1}{2}\left(1-\cos\phi_i\right)\end{array} \tag{9}$$

where $x,y=\uparrow$or$\downarrow$. Obviously, the probabilities of joint measurements in Eqs.(8) exhibit cosinusoidal modulations via the variable phase ϕ_i which yields two-particle interference fringes for the entangled state $|\Psi\rangle$, whereas there is no one-particle interference fringes. Conversely, a cosinusoidal dependence of the single probabilities on the variable phase ϕ_i in Eqs.(9) yields one-particle interference fringes for the product state $|\Phi\rangle$, but no genuine two-particle fringes, in that the variation of the joint probability $p\left(|x\rangle_1|y\rangle_2\right)$ results only from the variations of the single probabilities $p\left(|x\rangle_1\right)$ and $p\left(|y\rangle_2\right)$. This is the reason for using the "corrected" definition for two-particle fringe visibility. Further, Eqs.(8) shows $p\left(|x\rangle_1|y\rangle_2\right)\neq p\left(|x\rangle_1\right)p\left(|y\rangle_2\right)$, a manifestation of quantum nonlocality associated with the entangled state $|\Psi\rangle$. From the definitions of Eqs.(2) and (3), the theoretical outcomes of the visibilities are found to be $V_1=V_2=0, V_{12}=1$ for $|\Psi\rangle$ and $V_1=V_2=1, V_{12}=0$ for $|\Phi\rangle$.

The phenomenon can also be partially explained via the spirit of the 'which-way' (WW) interferometry experiment to test Bohr complementarity: any attempt to gain WW information unavoidably destroys the interference pattern [2,3]. For example, when the composite system of particles 1+2 is prepared in the entangled state $|\Psi\rangle$, WW information of one particle is stored in the states of the other through their correlation. Consequently, as far as a single particle (1 or 2) is concerned, there are full WW information and no one-particle fringes. However, for the pair of particles 1+2, there is no way to determine whether 1+2 takes the composite path $|\uparrow\rangle_1|\uparrow\rangle_2$ or the composite path $|\downarrow\rangle_1|\downarrow\rangle_2$, and hence perfect two-particle interference pattern displays. In contrast, if the system is in the product state $|\Phi\rangle$, it is impossible to determine the path of one particle through the other, and thus there is one-particle interference.

III. COMPLEMENTARITY IN THE GENERAL CASE

Now, we turn to the general case from two extreme $|\Psi\rangle$ and $|\Phi\rangle$. The most general state of pure two-particle source 1+2 that can be formed from the basis of $\{|\uparrow\rangle_i, |\downarrow\rangle_i\}$ is

$$|\Theta\rangle=\gamma_1|\uparrow\rangle_1|\uparrow\rangle_2+\gamma_2|\uparrow\rangle_1|\downarrow\rangle_2+\gamma_3|\downarrow\rangle_1|\uparrow\rangle_2+\gamma_4|\downarrow\rangle_1|\downarrow\rangle_2, \tag{10}$$

where

$$|\gamma_1|^2+|\gamma_2|^2+|\gamma_3|^2+|\gamma_4|^2=1, \tag{11}$$

where $\gamma_i=|\gamma_i|e^{i\delta_i}$ are complex numbers. As the visibilities are independent of the phases δ_i, in our analysis, all of γ_i are taken to be real numbers [26].

Likewise, the unitary operation U in Eq.(7) is applied to $|\Theta\rangle$ and straightforward calculations yield

$$\begin{array}{l} p\left(|\uparrow\rangle_1\right)=\frac{1}{2}\left[1+2\left(\gamma_1\gamma_3+\gamma_2\gamma_4\right)\cos\phi_1\right],\\ p\left(|\downarrow\rangle_1\right)=\frac{1}{2}\left[1-2\left(\gamma_1\gamma_3+\gamma_2\gamma_4\right)\cos\phi_1\right],\\ p\left(|\uparrow\rangle_2\right)=\frac{1}{2}\left[1+2\left(\gamma_1\gamma_2+\gamma_3\gamma_4\right)\cos\phi_2\right],\\ p\left(|\downarrow\rangle_2\right)=\frac{1}{2}\left[1-2\left(\gamma_1\gamma_2+\gamma_3\gamma_4\right)\cos\phi_2\right],\end{array} \tag{12}$$

and

$$\overline{p}\left(|\uparrow\rangle_1 |\uparrow\rangle_2\right) = \frac{1}{4}\left[1 + M\cos\phi_1\cos\phi_2 - N\sin\phi_1\sin\phi_2\right] \tag{13}$$

where

$$\begin{array}{l} N = 2\left(\gamma_1\gamma_4 - \gamma_2\gamma_3\right) \\ M = 2\left(\gamma_1\gamma_4 + \gamma_2\gamma_3\right) - 4\left(\gamma_1\gamma_3 + \gamma_2\gamma_4\right)\left(\gamma_1\gamma_2 + \gamma_3\gamma_4\right) \end{array}, \tag{14}$$

and the similar expressions for other $\bar{p}\left(|x\rangle_1 |y\rangle_2\right)$. It is readily verified that $|N| \geqslant |M|$ by using the normalization condition of Eq.(11). With the derivative method [26], the maximal and minimal values of $\overline{p}\left(|\uparrow\rangle_1 |\uparrow\rangle_2\right)$ are deduced as: $\overline{p}_{\text{max,min}}\left(|\uparrow\rangle_1 |\uparrow\rangle_2\right) = \frac{1}{4}\left(1 \pm |N|\right)$, which can be achieved only when $\left(\cos\phi_1\cos\phi_2, \sin\phi_1\sin\phi_2\right) = (0, \pm 1)$, i.e., $\left(\phi_1, \phi_2\right) = \left(n\pi + \pi/2, m\pi + \pi/2\right), (n, m = 0, \pm 1, ...)$. Hence, on substituting for the maximal and minimal values of these probabilities in Eqs.(2) and (3), one gets

$$\begin{array}{l} V_1 = \left|2\left(\gamma_1\gamma_3 + \gamma_2\gamma_4\right)\right|, \\ V_2 = \left|2\left(\gamma_1\gamma_2 + \gamma_3\gamma_4\right)\right|, \\ V_{12} = \left|2\left(\gamma_1\gamma_4 - \gamma_2\gamma_3\right)\right|, \end{array} \tag{15}$$

and

$$V_i^2 + V_{12}^2 = 1 - \left[\left(\gamma_2^2 - \gamma_3^2\right) - (-1)^i\left(\gamma_1^2 - \gamma_4^2\right)\right]^2 \leq 1, \quad (i = 1, 2). \tag{16}$$

The complementarity relation is achieved for any pure state of a composite two-particle system. In this type of interference experiments, another interesting physical quantity is the WW information of one particle when the other serves as the WW maker. Because the two complementarities — between path distinguishability and single-particle visibility and between one- and two-particle visibilities — are intimately connected, the one-particle visibility V_i enters in the same way in both of them [8]. Without any measurement, the available *a priori* WW knowledge of one particle is described by the predictability P_i of the alternatives, which represents Man's knowledge before measure; while the distinguishability D_i denotes the maximal information about the "path" that can be extracted from an appropriate measure, which is Nature's information about the actual alternative; usually $P_i \leq D_i$ [10]. For all pure states $|\Theta\rangle$, the complementarity of single particle [8–10]

$$V_i^2 + D_i^2 = 1 \tag{17}$$

holds, while

$$V_i^2 + P_i^2 \leq 1, \tag{18}$$

where, by definition, the predictability P_i of the path for particle i reads [8–10]

$$P_i = \left|w_{i\uparrow} - w_{i\downarrow}\right| = \left|\left(\gamma_2^2 - \gamma_3^2\right) - (-1)^i\left(\gamma_1^2 - \gamma_4^2\right)\right| \tag{19}$$

where w_{ix} are the probabilities that particle i takes one way $|\uparrow\rangle$ or the other $|\downarrow\rangle$. Comparing inequality (16) with Eq.(19), an equality

$$V_i^2 + V_{12}^2 + P_i^2 = 1 \tag{20}$$

is achieved, which gives the relation among three physical quantities: V_i, V_{12} and P_i. In fact, Eq.(20) covers both of two complementarity relations of inequalities (16) and (18) and implies the condition of saturating the upper bound of the two inequalities. For example, if the state of particle i is prepared symmetrically ($w_{i\uparrow} = w_{i\downarrow} = \frac{1}{2}$) so that $P_i = 0$, i.e., there is nothing predictable about the paths in a symmetric one-particle counterpart, the one- and two-particle complementarity will be of the form of an equality. Or otherwise, if particle i is an asymmetric state ($w_{i\uparrow} \neq w_{i\downarrow}$), that is, one path is more likely than the other to begin with $P_i > 0$, the complementarity is strictly limited in the inequality (16), because partial WW information P_i restrains full one-particle interference visibility V_i.

As the complementarity is tightly associated with the property of the composite system, it would be natural to examine the relationships between these physical quantities and the entanglement E of the system. By calculating the von Neumann entropy [28], the entanglement E for the pure state $|\Theta\rangle$ in Eq.(10) goes as

$$E=-\frac{1-\sqrt{1-V_{12}^2}}{2}\log_2\left(\frac{1-\sqrt{1-V_{12}^2}}{2}\right)-\frac{1+\sqrt{1-V_{12}^2}}{2}\log_2\left(\frac{1+\sqrt{1-V_{12}^2}}{2}\right). \tag{21}$$

As illustrated in Fig. 2, this monotonically increasing relation between the two-particle visibility V_{12} and the entanglement E indicates the visibility of two-particle interference fringes V_{12} is entirely governed by the entanglement E of pure two-particle source: the absence of entanglement will results in zero-visibility two-particle interference fringes (e.g., the $|\Phi\rangle$ state in Eq.(5), $E=0$, $V_{12}=0$), and the prefect entanglement, the full two-particle interference fringes (e.g., the $|\Psi\rangle$ state in Eq.(4), $E=1$, $V_{12}=1$). From Eq.(21) one cannot directly make out clear relations between V_i or D_i and E. However, from Eq.(20) and Eq.(17), $D_i^2=P_i^2+V_{12}^2$ is gained, which means that D_i contains both of the *a priori* WW information P_i and the additional information V_{12} encoded in the entanglement between the object and the WW maker due to the entire determination of V_{12} by E. Since *a priori* knowledge P_i only lies on the feature of initial single subsystem rather than that of the composite system, these quantities can be studied with a given *a priori* WW knowledge P_i. For instance, in a relevant symmetric one-particle interferometer ($P_i=0$), then $V_i=\sqrt{1-V_{12}^2}$ and $D_i=V_{12}$, which tie up with the entanglement E (see Fig.2): the entanglement E is enhanced, more and more Nature's WW information is stored in the states of the marker particle through their correlation so that D_i rises whereas the one-particle visibility decreases, as discussed and experimentally testified in Ref. [22]. Most of one-particle interference experiments [13,19,20,22] are discussed in this kind of symmetric interferometer. If the interferometer begins with $P_i>0$, V_i and D_i still bear the similar dependence on the entanglement E, this monotonically decreasing relation for V_i and this monotonically increasing relation for D_i except for the wholly alterative amplitudes based on *a priori* knowledge P_i, while V_{12} does not alter the relation on E at all. Consequently, this figure also illuminates an important view that quantum correlation is completely responsible for complementarity.

In order to discuss these relations in greater detail, we study some special families of states. When $|N|=|M|$, the following two situations can be distinguished:

1) $\gamma_1\gamma_4=\gamma_2\gamma_3$: $|\Theta\rangle$ corresponds to any product state ($E=0$), then $V_{12}=0$. When $P_i=0$ (i.e., $\gamma_1=\gamma_4=\gamma_2=\gamma_3=\pm\frac{1}{2}$), we recover the $|\Phi\rangle$ state of Eq.(5) (the sign makes no difference on these physical quantities), $V_i=1$. When $P_i>0$, the one-particle interference visibility is limited by $V_i^2+P_i^2=1$.

2) $\gamma_2=\gamma_3=0$ or $\gamma_1=\gamma_4=0$: $|\Theta\rangle=\gamma_1|\uparrow\rangle_1|\uparrow\rangle_2+\gamma_4|\downarrow\rangle_1|\downarrow\rangle_2$ or $|\Theta\rangle=\gamma_2|\uparrow\rangle_1|\downarrow\rangle_2+\gamma_3|\downarrow\rangle_1|\uparrow\rangle_2$, which are entangled. When $P_i=0$, (i.e., $\gamma_1=\gamma_4=\pm\frac{1}{\sqrt{2}}$ or $\gamma_2=\gamma_3=\pm\frac{1}{\sqrt{2}}$), we retrieve the $|\Psi\rangle$ state of Eq.(4) or the similar state with $|\uparrow\rangle_2$ and $|\downarrow\rangle_2$ interchanged which are the maximal entangled states ($E=1$); in either case $V_i=0$ and $V_{12}=1$. When $0<P_i<1$, ($\gamma_1\neq\gamma_4$ or $\gamma_2\neq\gamma_3$), the $|\Theta\rangle$ state is partially entangled ($0<E<1$), it then follows $0<V_{12}<1$ which is limited by $V_{12}^2+P_i^2=1$ in the intermediate regime, but $V_i=0$ and $D_i=1$, the "path" can still be completely determined by a suitable measurement. When $P_i=1$ (one of two γ_i in $|\Theta\rangle$ to be zero), the $|\Theta\rangle$ state again recovers a product state ($E=0$), $V_{12}=0$ and $V_i=0$.

The cases discussed above are symmetric for particle 1 and particle 2 ($P_1=P_2$). We continue on investigating a special asymmetric case ($P_1\neq P_2$):

3) When $|\Theta\rangle$ is of this form

$$|\psi(\alpha,\beta)\rangle=\frac{1}{\sqrt{2}}\left[|\uparrow\rangle_1(\cos\alpha|\uparrow\rangle_2+\sin\alpha|\downarrow\rangle_2)+|\downarrow\rangle_1(\cos\beta|\uparrow\rangle_2+\sin\beta|\downarrow\rangle_2)\right], \tag{22}$$

In this case, $P_1=0$, $P_2=|\cos(\alpha+\beta)\cos(\alpha-\beta)|$. Simple substitutions produce $V_1=|\cos(\alpha-\beta)|$, $V_2=|\sin(\alpha+\beta)\cos(\alpha-\beta)|$ and $V_{12}=|\sin(\alpha-\beta)|$, which result in $V_1^2+V_{12}^2=1$ whereas $V_2^2+V_{12}^2\leq 1$. However, whether for particle 1 or for particle 2, $V_i^2+V_{12}^2+P_i^2=1$ is satisfied. If $\alpha-\beta=\frac{\pi}{2}$, we again turn to the state with $P_1=P_2=0$:

$$|\psi(\alpha)\rangle=\frac{1}{\sqrt{2}}\cos\alpha\left[|\uparrow\rangle_1|\uparrow\rangle_2+|\downarrow\rangle_1|\downarrow\rangle_2\right]+\frac{1}{\sqrt{2}}\sin\alpha\left[|\uparrow\rangle_1|\downarrow\rangle_2+|\downarrow\rangle_1|\uparrow\rangle_2\right]. \tag{23}$$

which was discussed in Ref. [26]: $V_i=|\sin(2\alpha)|$ and $V_{12}=|\cos(2\alpha)|$ which make $V_i^2+V_{12}^2=1$.

IV. EXPERIMENTAL INVESTIGATIONS

In our experiments, the chosen quantum system is ^{13}C-labeled chloroform $^{13}CHCl_3$ (Cambridge Isotope Laboratories, Inc.) molecules with the hydrogen nuclei (^{1}H) for particle 1 and the carbon nuclei (^{13}C) for particle 2. The spin-spin coupling constant J between ^{13}C and ^{1}H is 214.95 Hz. The relaxation times were measured to be $T_1=4.8$ *sec* and $T_2=3.3$ *sec* for the proton, and $T_1=17.2$ *sec* and $T_2=0.35$ *sec* for carbon nuclei. Experiments were performed on a BrukerARX500 spectrometer with a probe tuned at 125.77MHz for ^{13}C and at 500.13MHz for ^{1}H by using a conventional liquid-state NMR techniques.

The quantum ensemble is firstly prepared in a pseudo-pure state $|\psi_0\rangle = |\uparrow\rangle_1 |\uparrow\rangle_2$ from thermal equilibrium by applying line-selective pulses with appropriate frequencies and rotation angles as well as a consequent magnetic gradient pulse [29]. Different two-particle source which corresponds to any $|\Theta\rangle$ state in Eq.(10) can be prepared from the $|\psi_0\rangle$ state by appropriate operatations, e.g., the entangled state $|\Psi\rangle$ of Eq.(4), the product state $|\Phi\rangle$ of Eq (5) and the special family of states $|\psi(\alpha)\rangle$ of Eq.(23) by applying, respectively, the following NMR pulse sequences: $Y_1(-\frac{\pi}{2})X_1(-\frac{\pi}{2})Y_1(\frac{\pi}{2})X_2(-\frac{\pi}{2})Y_2\left(\frac{\pi}{2}\right)J_{12}(\frac{\pi}{2})Y_2(\frac{\pi}{2})$, $Y_1(\frac{\pi}{2})Y_2(\frac{\pi}{2})$ and $Y_2\left(\frac{\pi}{2}\right)X_2(\frac{\pi}{2})J_{12}(\frac{\pi}{2}-2\alpha)X_2\left(-\frac{\pi}{2}\right)Y_1(\frac{\pi}{2})$ to be read from left to right. Here $Y_i(\theta)$ denotes an θ rotation about $\hat{y}$ axis on particle i and so forth, and $J_{12}(\varphi)$ represents a time evolution of $\varphi/\pi J_{12}$ under the scalar coupling between spins 1 and 2. Then the transformation $U_i(\phi_i)$ to achieve the operations of phase shifters and beam splitters was realized by the NMR pulse sequence $X_i(-\theta_1)\,Y_i(\theta_2)\,X_i(-\theta_1)$ with $\theta_1 = \tan^{-1}(-\sin\phi_i)$ and $\theta_2 = 2sin^{-1}(-cos\phi_i/\sqrt{2})$. $U_i(\phi_i)$ with a set of appropriate values θ_1 and θ_2 was repeatedly applied in experiments to vary the value of ϕ_i from 0 to 2π. Finally, the probabilities of single and joint measurements were obtained by quantum state tomography [30] to reconstruct the diagonal elements of the output density matrices, which can be completed by employing reading-out pulses $Y_1(\frac{\pi}{2})$ and $Y_2(\frac{\pi}{2})$ after a gradient pulse field to record ^{1}H and ^{13}C spectra, respectively. The quantitative measurements of the one- and two-particle interference visibilities were gained by monitoring the variations of these probabilities versus ϕ_i. In our experiments, the complementarity test is restricted in the cases of $P_i = 0$, that is, to verify $V_i^2 + V_{12}^2 = 1$.

For two extreme cases, we simultaneously varied the values of ϕ_1 and ϕ_2 with the respective increment of $\pi/16$. Applying the procedure stated above, the experimental results are shown in Fig. 3. As expected, for the entangled state $|\Psi\rangle$, two-particle interference fringes are displayed (Fig. 3(b)) but almost no one-particle interference fringes (Fig. 3(a)). and the opposite situation for the product state $|\Phi\rangle$ (Fig. 3(c) and (d)). From these experimental data points in Fig. 3, the measured values of the visibilities were extracted: $V_1 = 0.12$, $V_2 = 0.14$, $V_{12} = 0.87$ for the entangled state $|\Psi\rangle$ and $V_1 = 0.93$, $V_2 = 0.99$, $V_{12} = 0.10$ for the product state $|\Phi\rangle$. By contrast with the theoretical expectations (see Sec II), more experimental errors were introduced in the case of $|\Psi\rangle$ inasmuch as the preparation of the entangled state $|\Psi\rangle$ was more complicated than that of the product state $|\Phi\rangle$.

The interferometric complementarity of the special family of states $|\Psi(\alpha)\rangle$ in the intermediate regime was testified with a similar procedure. As $\overline{p}(K_1K_2)$ is a function of ϕ_1 and ϕ_2 whose extrema $\overline{p}_{\max,\min}(K_1K_2)$ were found at $(\phi_1, \phi_2) = \left(n\pi + \frac{\pi}{2}, m\pi + \frac{\pi}{2}\right)$ in the theoretical calculations, we scanned one of ϕ_i while fixing the other into $\pi/2$ for a given state $|\Psi(\alpha)\rangle$, then repeated experiment with interchanging them, instead of simultaneously scanning ϕ_1 and ϕ_2. The procedure was repeated for different α. For the convenience of experiments, we changed the values of α from $\pi/4$ to $21\pi/16$ with the increment of $\pi/16$. From the desirable interference fringes shown from variations of the normalized populations versus ϕ_i, the visibilities were obtained. The measured and theoretical visibilities of one- and two-particle interference $V_1(\alpha)$, $V_2(\alpha)$ and $V_{12}(\alpha)$ are plotted in Fig. 4(a), together with entanglement $E(\alpha)$ of the two-particle system. These experimental results are in good agreement with the theoretical expectations. This figure also shows the relationship between the visibilities and entanglement, i.e., $E(\alpha)$ varies synchronously with $V_{12}(\alpha)$ and has the opposite variation trend with $V_i(\alpha)$, consistent with the theoretical analyses in Sec.III. The experimental data of the complementarity relations for $V_i^2(\alpha) + V_{12}^2(\alpha)$ are also depicted in Fig. 4(b). The initial state of two-particle source is a symmetric state with no *a priori* WW knowledge of particle 1 and 2 ($P_1 = P_2 = 0$), so that all data in Fig. 4(b) should be unity.

In these experiments, the estimated errors are less than $\pm 10\%$ due to the inhomogeneity of the RF field and static magnetic field, imperfect calibration of RF pulses and signal decay during the experiments. If we take into account the imperfections of the experiments, the measured data in our NMR experiments agree well with theory.

V. CONCLUSION

For a general pure two-particle source, we derive the interferometric complementarity between one- and two-particle interference visibilities and obtain an equality among the one-particle interference visibility V_i, two-particle interference visibility V_{12} and the predication P_i of the path of single particle: $V_i^2 + V_{12}^2 + P_i^2 = 1$. The equality not only embodies two complementarity relations of $V_i^2 + V_{12}^2 \leq 1$ and $V_i^2 + P_i^2 \leq 1$, but also implies the conditions of their saturating the upper bound of them, e.g., for the one- and two-particle interference, the duality of $V_i^2 + V_{12}^2 = 1$ holds only when the interferometer involves a symmetric single-particle counterpart (i.e., $P_i = 0$). In fact, the equality is also a demenstration of the relationships among one-paritcle wave-like (V_i), particle-like (P_i) and two-particle wave-like (V_{12}) attributes. In addition, we also manifest the role of entanglement in this type of interference experiments, which reveals whether in a symmetric way or in an asymmetric one, the greater entanglement is the quantum composite system, the lower the visibility of two-particle interference fringes whereas the higher the visibility of two-particle interference fringes. Meanwhile, we also consider the relation between two interferometric complementarities. The one- and two-particle interference complementarity has been testified for a kind of special symmetric interferometer ($P_1 = P_2 = 0$) in a spin NMR ensemble, including two extreme cases and a special family of states. The experimental results showed the theoretical predictions of quantum mechanics within the experimental errors. Though the test was performed over interatomic distances, the experimental scheme originates from quantum version and their dynamical evolution is quantum mechanical. Therefore, these experiments also indicate that non-classical properties associated with entanglement can be displayed in NMR ensemble as long as the initial state is well prepared.

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Figure Captions

Fig. 1 Schematic two-particle interferometer using beam splitters BS_1, BS_2 and phase shifters ϕ_1, ϕ_2.

Fig. 2 The entanglement E versus visibility V_{12} for pure two-particle source (denoted by the solid line), visibility V_i and distinguishability D_i of a single particle in different *a priori* WW knowledge P_i: the dotted line of V_i and the solid line of $D_i = V_{12}$ for $P_i = 0$; the dashed line of V_i and the dashdotted line of $D_i = V_{12}$ for $P_i = 0.4$.

Fig. 3 The single and "corrected" joint probabilities detected in the one- and two-particle interference, (a) and (b) for the entangled state $|\Psi\rangle$, (c) and (d) for the product state $|\Phi\rangle$. In (a) and (c), data points $\Box$, $+$, $\bigcirc$ and $\times$ denote the single probabilities $p\left(|\uparrow\rangle_1\right), p\left(|\downarrow\rangle_1\right), p\left(|\uparrow\rangle_2\right)$ and $p\left(|\downarrow\rangle_2\right)$, respectively; in (b) and (d), $\bigcirc$, $\times$, $\Box$ and $+$ denote the "corrected" joint probabilities $\overline{p}\left(|\uparrow\rangle_1|\uparrow\rangle_2\right), \overline{p}\left(|\uparrow\rangle_1|\downarrow\rangle_2\right), \overline{p}\left(|\downarrow\rangle_1|\uparrow\rangle_2\right)$ and $\overline{p}\left(|\downarrow\rangle_1|\downarrow\rangle_2\right)$, respectively. Theoretical curves are depicted with the solid lines.

Fig. 4 The visibilities and the complementarity relation of a special family of states $|\psi(\alpha)\rangle$ as a function of the angle α. (a) Data points $\bigcirc$, $+$ and $*$ denote, respectively, the visibilities of one-particle interference V_1, V_2 and two-particle interference V_{12}. The solid lines are the theoretical expectations and the dotted line denotes the theoretical curve of entanglement E. (b) Experimental test of the complementarity relation $V_1^2 + V_{12}^2$ (denoted by data ponits $\Box$) and $V_2^2 + V_{12}^2$ (denoted by data ponits $\triangledown$) are plotted as a function of α. The solid line represents the theoretical expectations.